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13. ABSTRACT (Maximum 200 words)

The objective of the present program is to study the structure and response of steady and unsteady laminar premixed and nonpremixed flames in reduced and elevated pressure environments through (a) non-intrusive experimentation, (b) computational simulation using detailed flame and kinetic codes, and (c) asymptotic analysis with reduced kinetic mechanisms. For the three year period supported by the grant, the following projects were completed and reported in nineteen journal-class articles: (1) Studies on unsteady flames; (2) Studies on high-pressure flames; (3) Re-examination of the counterflow technique in flame speed determination; (4) Asymptotic structure of premixed methane/air flames; (5) Theories on flame extinction; (6) Numerical nlgorithm for generating S-curves; (7) Thermophoretic effects on seeding particles in LDV measurements of flames; and (8) Review activities.

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SUMMARY/OVERVIEW

The objective of the present program is to study the structure and response of steady and unsteady laminar premixed and nonpremixed flames in reduced and elevated pressure environments through (a) non-intrusive experimentation, (b) computational simulation using detailed flame and kinetic codes, and (c) asymptotic analysis with reduced kinetic mechanisms. For the three year period supported by the grant, the following projects were completed and reported in nineteen journal-class articles:

1. Studies on unsteady flames
2. Studies on high-pressure flames
3. Re-examination of the counterflow technique in flame speed determination
4. Asymptotic structure of premixed methane/air flames
5. Theories on flame extinction
6. Numerical nlgorithm for generating S-curves
7. Thermophoretic effects on seeding particles in LDV measurements of flames
8. Review activities

Specific interests and accomplishments of these projects are briefly described in the next section.

ACCOMPLISHMENTS

1. Studies on Unsteady Flames

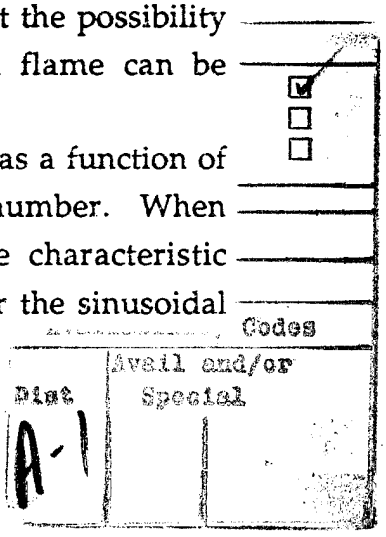
A crucial influence on the flame behavior which so far has not been adequately addressed is the effect of unsteadiness of the environment on the flame behavior. This issue is of particular relevance to the modeling of turbulent flames through the concept of laminar flamelets. These flamelets are subjected to fluctuating flows with various intensities of straining, and it is reasonable to expect that the flame would respond differently in an oscillating strained flow field than in a steady strained flow field.

During the program period we have first analyzed the structure and response of a one-dimensional chambered flame in an oscillating flow field with small

amplitudes of fluctuation, using large activation energy asymptotics. In particular, we are concerned with the situation in which the characteristic time for flow unsteadiness is comparable with that of convection and diffusion, yet much larger than that of chemical reaction. In such cases, flames can be described as quasi-steady reactive layers embedded in, and interacting with, time-varying non-reactive transport zones. Our analytical results show that the deviation from the mean flame location and temperature decreases as the frequency of flow oscillation increases. Thus, as the frequency increases, flow unsteadiness becomes extremely rapid such that the flame cannot readily respond in a quasi-steady manner, i.e. the flame is hardly affected by the unsteady flow in the high frequency limit. By further analyzing the reaction zone structure, we have assessed the effect of the unsteady flow field on the extinction characteristics of the flame.

The above chambered flame analysis was subsequently extended to a counterflow diffusion flame subjected to an oscillating strain rate. The characteristic oscillation time of practical interest is found to be of the same order as the characteristic diffusion time of the flame, so that the flame structure again consists of a quasi-steady reactive-diffusive layer embedded in the outer unsteady-convective-diffusive zones. A linear analysis is conducted by assuming that the amplitude of the strain rate oscillation is small relative to the mean strain rate. It has been found that the flame response is controlled mainly by two effects: (a) the response of the convective mass flux into the reaction sheet, which is directly related to the flow-field variation applied at the boundary, and (b) the response of the reaction sheet to adjust the reduced residence time due to finite-rate chemistry. For flames near equilibrium, the former effect tends to be dominant, so that the response of the net heat release is in phase with the strain rate oscillation. For flames near extinction, however, the finite-rate chemistry effect overtakes the fluid-dynamic effect such that increasing strain rate leads to a reduction of the reactivity of the flame during the oscillatory cycle. As such, the net heat release response of the near-extinction flame becomes out of phase with the strain rate oscillation in the sense of the Rayleigh's criterion. Results of the present study suggest the possibility that the unsteady characteristics of the near-extinction diffusion flame can be significantly different from those in the Burke-Schumann limit.

The above study was then extended to an *arbitrary* strain rate as a function of time, with the assumptions of constant density and unity Lewis number. When specialized to the case of an impulsively-applied strain rate, the characteristic relaxation time as well as the extinction delay time are derived. For the sinusoidal



strain rate, the reaction sheet oscillation is found to be asymmetrical with respect to its initial location, protruding more toward the freestream side. Furthermore, the amplitude of the oscillation decreases with frequency, and its phase delay approaches $\pi/2$ in the high frequency limit, as observed in previous experimental and numerical studies. For a given amplitude of oscillation, the flame is more easily extinguished when the characteristic time of oscillation is sufficiently long. When this result is applied to the current understanding of turbulent flames, it suggests that the laminar flame sheet can be sustained at higher Reynolds numbers. This is because there exists a range of eddies which, while possessing a sufficiently large strain rate to extinguish the flame in the steady limit, do not have sufficiently long characteristic time to effect extinction. Thus it seems reasonable to suggest that the applicable range of the laminar flamelet regime may be wider than can be expected from quasi-steady considerations.

Experimentally, we have constructed a counterflow burner with strain rate oscillations being applied by loud speakers. An important consideration here is the ability to independently vary the frequency and amplitude of the oscillation. Preliminary results seem to indicate that, for a given frequency, extinction occurs at a constant maximum strain rate regardless of the steady-state strain rate. This would imply that extinction is a quasi-steady process, with the reaction zone only affected by the instantaneous strain rate it experiences. Unsteadiness, however, does seem to have a second order effect in that the instantaneous extinction strain rate is found to increase with frequency.

2. Studies on High-Pressure Flames

We have previously performed extensive measurements of the laminar flame speeds of a variety of hydrocarbon fuels with air for pressures up to three atmospheres. Through these studies we have come to recognize the significance of the system pressure in influencing chemical kinetics in general and the flame burning rate in particular. For example, in combustion modeling with the simplified kinetics of one-step overall reaction, the pressure exponent n is usually assumed to be a constant which is also frequently taken to be two for a second order reaction. Our results, however, have shown that not only n is not a constant, but it invariably is less than two. Specifically, it is close to one for the stoichiometric burning of methane in atmospheric air. Furthermore, it decreases with increasing pressure and inert dilution. In a highly-diluted situation it can even assume negative values, indicating that the flame burning intensity decreases with

increasing pressure. Such behavior can be explained on the basis of the relative efficiencies of the two-body high-activation-energy, temperature-sensitive branching reactions versus the three-body, low-activation-energy, temperature-insensitive termination reactions.

Recognizing the importance of pressure on the flame behavior, and that practical aero-engines do operate under pressures much higher than those studied, we have constructed a high-pressure burner with an operational limit of 15 atmospheres. We have subsequently conducted experimentations on the extinction of counterflowing methane/air diffusion flames up to 8 atmosphere. The kinetic structure of the flame is currently being studied.

Recent interests in high-speed aero-propulsion have also led to considerable research on hydrogen/oxygen chemistry, as well as its coupling to fluid flows. Because of the high-speed nature of the flow, the available residence time for mixing and chemical reaction is significantly reduced, implying that it is of importance to understand the ignition and extinction phenomena involving hydrogen/oxygen mixtures under variable pressures. We have thus measured the local extinction strain rates of nonpremixed counterflow flames of diluted hydrogen against air, at pressures of 0.5 to 1.0 atmosphere. The measured data compare well with results obtained from computational simulation with detailed chemistry and transport. We have also performed additional computational studies of the pressure effect on flame extinction. The relationship of extinction temperatures with pressure for this system was found to exhibit the familiar non-monotonic "Z" shaped dependency as observed for the homogeneous hydrogen/oxygen explosion limits. This implies that an increase in pressure could render a mixture to change from an extinguished state, to a burning state, and back to an extinguished state. This behavior is explained on the basis of the intrinsic chain branching-termination kinetics of hydrogen oxidation.

3. Re-Examination of the Counterflow Technique in Flame Speed Determination

The accuracy of the laminar flame speed determination by using the counterflow twin flame technique has been computationally and experimentally examined in light of the recent understanding that linear extrapolation of the reference upstream velocity to zero strain rate would yield a value higher than that of the laminar flame speed, and that such an over-estimate can be reduced by using either lower strain rates and/or larger nozzle separation distances. A systematic evaluation of the above concept has been conducted and verified for the ultra lean

hydrogen/air flames which have relatively large Karlovitz numbers, even for small strain rates, because of their very small laminar flame speeds. Consequently, the significantly higher values of the previous experimentally measured flame speeds, as compared to the independently calculated laminar flame speeds, can now be attributed to the use of nozzle separation distances which were not sufficiently large and/or strain rates which were not sufficiently small. Thus by using lower strain rates and larger nozzle separation distances the experimentally and computationally re-determined values of these ultra lean hydrogen/air flames agree well with the calculated laminar flame speeds. The laminar flame speeds of methane/air and propane/air mixtures have also been experimentally re-determined over extensive ranges of the equivalence ratio and are found to be slightly lower than previously reported experimental values.

4. Asymptotic Structure of Premixed Methane/Air Flames

In many of the previous flame studies using reduced mechanisms, the activation energies of the individual elementary reactions are not considered to be large enough to employ large activation energy asymptotics. Instead, the reaction rates are assumed to have a power law dependence on temperature, and the ratio of the individual reaction rates are used as small parameters in the so-called rate-ratio asymptotics. Since the power law dependence on temperature does not exhibit the same kind of sensitivity to temperature variations as the Arrhenius approximation, the response of the flame to various external perturbations is not very significant. Consequently, analyses of this kind have yielded results which are not completely satisfactory. For example, for premixed methane/air flames, it was found that the Damköhler number from the leading order flame structure analysis does not depend on the burning rate, and therefore needs to be determined in a somewhat *ad hoc* manner. Furthermore, the key termination reaction was found to be the propagation step rather than the three-body recombination reaction.

In this study we have analyzed the structure of premixed methane/air flames, using the four-step reduced mechanism of Peters and Williams, but retaining the Arrhenius form for the individual reactions except for the three-body recombination reaction which has zero activation energy. Thus activation energy asymptotics can still be applied and a complete description of the flame structure is provided. The role of each of the overall reaction steps is determined, and the key termination reaction is found to be the three-body recombination reaction. The results include the evaluation of the burning rate in terms of pressure, equivalence

ratio, and a parameter that represents the ratio of the competing rates of the branching and termination reactions. In particular, we find that when this rate-ratio is reduced to a critical value, the burning rate decreases to zero and the flame "extinguishes". For the opposite limit that the rate-ratio is large, we find that the burning rate asymptotes to a constant value, and the problem is governed by that of the premixed flame regime of Liñán. Consequently, we are able to adopt his correlation curve as an approximate expression for the burning rate in this limit.

5. Theories on Flame Extinction

A classical model problem for the study of premixed flame extinction is that of Spalding, who analyzed a one-dimensional freely-propagating flame with a temperature-sensitive one-step overall reaction and radiative heat loss. More sophisticated analyses have since been performed using one-step chemistry and activation energy asymptotics. These studies predict that extinction occurs when the ratio of the burning rate to its adiabatic value is reduced to $e^{-1/2} \approx 0.61$, a result that is insensitive to the nature of the heat loss. Subsequent numerical computations that consider detailed transport and chemistry also predict extinction when the burning rate is reduced to roughly 60% of its adiabatic value, suggesting that this value may represent a universal constant, independent of reaction and loss mechanisms.

In order to gain a better understanding of the role of dominant kinetic parameters, we have revisited this classical problem and have performed asymptotic analyses with multi-step reaction mechanisms. We first employed the two-step Zel'dovich-Liñán mechanism which consists of a branching reaction and a competing recombination reaction, thus capturing the chain nature of real flames. The analysis again predicts the normalized burning rate at extinction to be $e^{-1/2}$.

We have also made considerable progress in understanding the extinction characteristics of nonadiabatic methane/air flames based on the reduced reaction mechanism of Peters and Williams. Extinction conditions were found explicitly in terms of these parameters. Results show that the critical value of the burning rate at extinction, though not a universal constant, varies only slightly, in the range between 0.61 and 0.64. It was found that at the (extinction) turning point, the burning rate is nearly the same for all curves, but the critical value of the heat loss parameter is seen to be less than that for one-step models. The corresponding reduction in flame temperature needed for extinction is therefore substantially less than that predicted by one-step models.

6. Numerical Algorithm for Generating S-Curves

Most combustion phenomena are intrinsically nonlinear because of the associated chain mechanisms and Arrhenius kinetics. Consequently, theoretical studies of their steady-state behavior frequently yield multiple solutions connected by turning points when a system response is plotted versus an imposed system parameter. A prominent example is the S-curve representation of quasi-one-dimensional strained flames which results when a flame response such as its maximum temperature is plotted versus the system strain rate. Specifically, the lower branch of the S-curve represents weakly-reactive states, the middle branch unstable solutions, and the upper branch vigorously-burning states. As such, the lower and upper turning points can be respectively defined as the ignition and extinction states of the system. Identification and description of these critical states of transition therefore commands both fundamental and practical interest.

When numerical solution is sought for the S-curve response, especially with detailed chemistry, difficulty is encounters as one attempts to continue the calculation from, say, the upper branch to the middle branch by negotiating the extinction turning point because the Jacobian matrix used in the Newton method becomes singular at the turning point. Consequently, the arclength continuation method has been applied to trace through the ignition and extinction turning points in a number of studies on strained flames.

While the arclength continuation method is clearly a valuable approach in generating the S-curve, special skill apparently is still needed in its implementation and as a result its use has not been wide spread. In this study we have developed a new, flame-controlling continuation method. Using the counterflow premixed and diffusion flames as examples, the method capitalizes on the distinct nature of the profile and location of the scalars of the flame properties, such as the temperature and species concentrations, in response to changes in the flow strain rate. Thus instead of using the strain rate as an imposed parameter and the scalars as the flame responses, the values of a flame scalar at a give location is used as an internal boundary condition while the strain rate becomes the flame response. The method appears to be fairly simple in implementation and efficient and robust in execution, especially in negotiating the turning points.

7. Thermophoretic Effects on Seeding Particles in LDV Measurements of Flames

In a recent experimental and computational study on the detailed dynamic, thermal, and chemical structure of adiabatic, laminar counterflow premixed flames,

we noticed that while close quantitative agreement between the measured and computed results exists for the scalar structure of the flame, the LDV-measured axial velocity profile consistently lags the calculated values by substantial amounts in the preheat zone of the flame. Order of magnitude estimates showed that such a lag could be due to the influence of thermophoresis on the LDV seeding particles in the high-temperature-gradient environment of the preheat zone. Indeed, when detailed calculations were performed for the motion of the seeding particles, under the influence of drag and thermophoresis and by using the computationally-determined flame structure to evaluate the various transport coefficients, the computed particle trajectory agreed well with the measured LDV velocity profile.

Since temperature increases monotonically in the direction of the flow in an adiabatic *premixed* flame, the effect of thermophoresis can be readily visualized. However, for a counterflow *diffusion* flame, the temperature peaks in the flow field such that the thermophoretic force acts in opposite direction in the fuel and oxidizer sides of the flame. Furthermore, since the flow is uni-directional in crossing the flame, and the direction also switches when the flame moves across the stagnation surface, the net dynamic response of the LDV particles can be very rich.

There are several implications of this finding. First, thermophoresis appears to be a factor that needs to be estimated and possibly accounted for when measuring flame properties and responses using LDV and PIV, especially for thin flames in local flow field of low convective velocities. Its potential influence on measuring the velocity statistics in turbulent flames also needs to be examined. Furthermore, caution is also needed in the study of flame chemistry by directly extracting the local temperature profile from the local LDV-velocity profile, without considering thermophoresis.

8. Review Activities

Two major review articles have been written on combustion phenomena whose understanding has been made possible through several long-term research programs including the present one. The first is on microgravity combustion in which interpretation of the flame structure and aerodynamic response reached fruition through the present program. The second is on the role of chain mechanisms in combustion phenomena in which chemical and aerodynamic effects on the flame structure and response are discussed from a unified viewpoint.

JOURNAL PUBLICATIONS

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12. "Opportunities and challenges of combustion in microgravity," by C.K. Law and G.M. Faeth, *Progress in Energy and Combustion Science*, Vol. 20, pp. 65-113 (1994).

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14. "Effects of pressure and dilution on the extinction of counterflow nonpremixed hydrogen-air flames," by P. Papas, I. Glassman and C.K. Law, *Twenty-Fifth Symposium (International) on Combustion*, pp. 1333-1339 (1994).

15. "Response of counterflow diffusion flames to oscillating strain rates," by H. G. Im, C. K. Law, J. S. Kim and F. A. Williams, *Combustion and Flame* , Vol. 100, pp. 21-30 (1995)

16. "Counterflow diffusion flames with unsteady strain rates," by H.G. Im, J.K. Bechtold and C.K. Law, *Combustion Science and Technology*, Vol. 106, pp. 345-361 (1995).

17. "The role of chain mechanisms in some fundamental combustion phenomena," by C.K. Law, *Physical and Chemical Aspects of Combustion*, in press.

18. "Further studies on effects of thermophoresis on seeding particles in measurements of strained flames," by C.J. Sung, J.S. Kistler, M. Nishioka and C.K. Law, *Combustion and Flame*, in press.

19. "A flame-controlling continuation method for generating S-curve responses with detailed chemistry," M. Nishioka, C.K. Law and T. Takeno, *Combustion and Flame*, in press.

RESEARCH PERSONNEL

H.G. Im (Doctoral student, graduated and currently Post-Doctoral Fellow at the Stanford/Ames Center for Turbulence Research)

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SIGNIFICANT INTERACTIONS

The laminar flame speed data that have been experimentally determined in the P.I.'s laboratory have been extensively adopted by chemical kineticists in their modeling efforts. In particular, the GRI (Gas Research Institute) Mechanism on methane oxidation used our data as its pivotal calibration point.

The theory of flammability limits is also being applied on a first principle basis to predict the flammability limits of practical combustible mixtures. At the latest it is being used by researchers at Sandia, NIST, and DuPont Chemicals.

INVENTIONS

None.